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The Use of Reciprocity in Atmospheric Source Inversion Problems

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Notation

Vectors and tensors are bold sans-serif characters (e.g., x, v). Operators are in calligraphic font (e.g., $\mathcal{L}$, $\mathcal{O}$). Integers representing cardinality of a particular set are capitalized using a "typewriter" font type (e.g., $\mathtt{N}$, $\mathtt{M}$). Standard non-capitalized, non-bold symbols represent simple scalars or scalar variables (e.g., a, b).

1. Motivation

The goal of the Event Reconstruction Project (Sugiyama et al., 2004) is to find the location and strength of atmospheric release points, both stationary and moving. Source inversion relies on observational data as input. The methodology is sufficiently general to allow various forms of data. In this report, we will focus on primarily on concentration measurements obtained at point monitoring locations at various times.

The algorithms being investigated in the Project are the MCMC (Markov Chain Monte Carlo), SMC (Sequential Monte Carlo) Methods, classical inversion methods, and hybrids of these. We refer the reader to the report by Johannesson et al. (2004) for explanations of these methods. These methods require computing the concentrations at all monitoring locations for a given "proposed" source characteristic (locations and strength history). It is anticipated that the largest portion of the CPU time will take place performing this computation. MCMC and SMC will require this computation to be done at least tens of thousands of times. Therefore, an efficient means of computing forward model predictions is important to making the inversion practical.

In this report we show how Green's functions and reciprocal Green's functions can significantly accelerate forward model computations. First, instead of computing a plume for each possible source strength history, we can compute plumes from unit impulse sources only. By using linear superposition, we can obtain the response for any strength history. This response is given by the forward Green's function.

Second, we may use the law of *reciprocity*. Suppose that we require the concentration at a single monitoring point x_m due to a potential (unit impulse) source that is located at x_s. Instead of computing a plume with source location x_s, we compute a "reciprocal plume" whose (unit impulse) source is at the monitoring locations x_m. The reciprocal plume is computed using a reversed-direction wind field. The wind field and transport coefficients must also be appropriately time-reversed (see following sections for actual details). Reciprocity says that the concentration of reciprocal plume at x_s is related to the desired concentration at x_m. Since there are many less monitoring points than potential source locations, the number of forward model computations is drastically reduced.

A separate important benefit of using Green's functions is that they allow us to succinctly characterize the linear dependence between the concentration field and the source strength field. This characterization allows us to exploit classical closed-form methods available for linear systems under gaussian statistical assumptions. These methods can yield highly appropriate proposal distributions for significantly speeding-up Monte Carlo algorithms for solving non-gaussian statistical problems. This approach is also likely to make the CPU costs of the Monte Carlo algorithm more predictable than purely Monte Carlo methods which have the possiblity to be highly unpredictable. This advantage is very important for "on-line" applications.

A good reference for Green's functions and the adjoint problem is Morse and Feschbach (1953), who derive the adjoint Green's function for the diffusion equation.

2. Source Inversion Problem for the Advective-Diffusion Equation

2.1. Mathematical Preliminaries

Instead of restricting ourselves to point sources, we will consider general source fields $s(\mathsf{x}, t)$.

Consider the concentration field in a domain Ω due to a release from $s(\mathsf{x}, t)$. The concentration is assumed to be a solution to the following partial differential equation for advective and dispersive transport with decay:

$$\frac{\partial c(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, t, \mathsf{v})c(\mathsf{x}, t) + \nabla \cdot [c(\mathsf{x}, t)\mathsf{v}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, \mathsf{v})\nabla c(\mathsf{x}, t) = s(\mathsf{x}, t), \tag{2.1}$$

for $\mathsf{x} \in \Omega$, $t \geq 0$. Note that, in general, the dispersion tensor k and the decay constant λ may depend locally on the wind velocity field $\mathsf{v} = \mathsf{v}(\mathsf{x}, t)$. We have used the notation $\mathsf{k}(\mathsf{x}, t, \mathsf{v})$ as a short-hand for $\mathsf{k}(\mathsf{x}, t, \mathsf{v}(\mathsf{x}, t))$, and $\lambda(\mathsf{x}, t, \mathsf{v})$ as a short-hand for $\lambda(\mathsf{x}, t, \mathsf{v}(\mathsf{x}, t))$. We require that $\mathsf{k}(\mathsf{x}, t, \mathsf{v})$ be a symmetric tensor that is positive definite, and $\lambda(\mathsf{x}, t, \mathsf{v})$ be a non-negative scalar field.

The wind field $\mathsf{v} = \mathsf{v}(\mathsf{x}, t)$ is assumed to be "incompressible":

$$\nabla \cdot \mathsf{v}(\mathsf{x}, t) = 0. \tag{2.2}$$

(It will be seen later that the above incompressibility assumption allows the "reciprocal problem" to be solved from a time-reversed version of the "forward problem". Otherwise, an additional source term is required in addition to the time-reversal.)

The source field $s(\mathsf{x}, t)$ is assumed to satisfy the condition:

$$\int_{-\infty}^{\infty} \int_{\Omega} s(\mathsf{x}, t)^2 \, \mathsf{dx} dt < \infty. \tag{2.3}$$

This condition rules out, among other things, sources that have infinite spatial extent and sources that have finite extent, but do not die off sufficiently fast enough in time. This condition is clearly satisfied for spatially finite sources that go to zero for $t > T$.

The solution to the above partial differential equation is assumed to satisfy an initial condition of zero concentration:

$$c(\mathsf{x}, t) = 0, \quad \mathsf{x} \in \Omega, \ t \leq 0, \tag{2.4}$$

and zero boundary condition on the boundary $\partial\Omega$ of Ω:

$$c(\mathsf{x}, t) = 0, \quad \mathsf{x} \in \Omega, \ t \geq 0. \tag{2.5}$$

For convenience, we may wish to take the outer boundary of the problem to be sufficiently far enough away that the concentrations at the boundary are negligible by the time the plume reaches there. Or, we may wish to extend our domain to infinity so that Ω becomes the entire three-dimensional space R^3.

We will restrict our solutions that have finite square integrals:

$$\int_{-\infty}^{\infty} \int_{\Omega} c(\mathsf{x}, t)^2 \, d\mathsf{x} dt < \infty, \tag{2.6}$$

and finite square integrals of their gradients:

$$\int_{-\infty}^{\infty} \int_{\Omega} |\nabla c(\mathsf{x}, t)|^2 \, d\mathsf{x} dt < \infty. \tag{2.7}$$

From the zero concentration boundary condition, the condition on the source term, and the diffusive nature of the partial differential equation, it can be shown that the solution $c(\mathsf{x}, t)$ satisfies

$$\lim_{t \to \infty} \int_{\Omega} c(\mathsf{x}, t)^2 \, d\mathsf{x} = 0. \tag{2.8}$$

That is, the mean-square concentration over the entire domain approaches zero as time goes to infinity.

2.2. The Advection-Dispersion Operator and Its Adjoint

We define the advection-dispersion operator, which we call $\mathcal{L}$, by

$$\mathcal{L}[\phi(\mathsf{x}, t)] \equiv \frac{\partial \phi(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, t, \mathsf{v})\phi(\mathsf{x}, t) + \nabla \cdot [\phi(\mathsf{x}, t)\mathsf{v}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, \mathsf{v})\nabla\phi(\mathsf{x}, t). \tag{2.9}$$

The resulting short-hand for (2.1) is

$$\mathcal{L}[c(\mathsf{x}, t)] = s(\mathsf{x}, t). \tag{2.10}$$

The operator $\mathcal{L}$ will be defined not only over functions ϕ that are concentration solutions $c(\mathsf{x}, t)$, but for any function ϕ that satisfies conditions (2.5) through (2.8). That is, ϕ must satisfy the following:

$$\phi(\mathsf{x}, t) = 0, \quad \mathsf{x} \in \Omega, \ \forall t, \tag{2.11}$$

$$\int_{\infty}^{\infty}\int_{\Omega}\phi(\mathsf{x},t)^2\,d\mathsf{x}dt<\infty,\qquad \int_{\infty}^{\infty}\int_{\Omega}|\nabla\phi(\mathsf{x},t)|^2\,d\mathsf{x}dt<\infty, \tag{2.12}$$

$$\lim_{t\to\infty}\int_{\Omega}\phi(\mathsf{x},t)^2\,d\mathsf{x}=0. \tag{2.13}$$

The "space-time inner product" between two functions ϕ and ψ is defined by

$$<\phi,\psi>\equiv\int_{-\infty}^{\infty}\int_{\Omega}\phi(\mathsf{x},t)\psi(\mathsf{x},t)\,d\mathsf{x}dt. \tag{2.14}$$

Now, given a linear operator $\mathcal{O}$ we define its *adjoint* operator as the operator $\mathcal{O}^t$ such that

$$<\mathcal{O}\phi,\psi>=<\phi,\mathcal{O}^t\psi>, \tag{2.15}$$

where both ϕ and ψ satisfy (2.11) through (2.13).

In Appendix A, it is shown that the adjoint operator $\mathcal{L}^t$ for the advection-dispersion operator $\mathcal{L}$ is given by

$$\mathcal{L}^t[\phi(\mathsf{x},t)]\equiv-\frac{\partial\phi(\mathsf{x},t)}{\partial t}+\lambda(\mathsf{x},t,\mathsf{v})\phi(\mathsf{x},t)-\nabla\cdot[\phi(\mathsf{x},t)\mathsf{v}(\mathsf{x},t)]-\nabla\cdot\mathsf{k}(\mathsf{x},t,\mathsf{v})\nabla\phi(\mathsf{x},t). \tag{2.16}$$

Note that $\mathcal{L}^t$ is defined only for functions ϕ satisfying (2.11) through (2.13).

An important note is that the adjoint operator $\mathcal{L}^t$ defined by (2.16) is of the same general form as the advection-dispersion operator $\mathcal{L}$ defined in (2.9) if we reverse time in all of the terms and reverse the wind field direction in the advective term. More details on this aspect will be given later.

2.3. The Green's Function for the Advection-Dispersion Equation

The Green's function $g_{\mathsf{x}'t'}(\mathsf{x},t)$ is a solution to (2.1) in the special case where the source field is a unit impulse of mass located at x', occurring in a single "instant" of time t'. That is, for the source function we write $s(\mathsf{x},t)=\delta_{\mathsf{x}'t'}(\mathsf{x},t)$, where the delta-"function" $\delta_{\mathsf{x}'t'}(\mathsf{x},t)$ is formally defined by

$$\int_{-\infty}^{\infty}\int_{\Omega}\delta_{\mathsf{x}'t'}(\mathsf{x},t)\phi(\mathsf{x},t)\,\mathsf{dx}dt=\phi(\mathsf{x}',t'), \tag{2.17}$$

for all integrable functions ϕ. An equivalent representation is $\delta_{\mathsf{x}'t'}(\mathsf{x},t)=\delta(\mathsf{x}-\mathsf{x}')\delta(t-t')$.

Substituting $s(\mathsf{x},t)=\delta_{\mathsf{x}'t'}(\mathsf{x},t)$ into (2.1), we have that for $t>t'$ the Green's function satisfies the equation:

$$\mathcal{L}[g_{\mathsf{x}'t'}(\mathsf{x},t)]=\delta_{\mathsf{x}'t'}(\mathsf{x},t). \tag{2.18}$$

For $t\le t'$ we require the initial condition:

$$g_{\mathsf{x}'t'}(\mathsf{x},t)=0,\ \ \mathsf{x}\in\Omega. \tag{2.19}$$

The following boundary condition is also required:

$$g_{\mathsf{x}'t'}(\mathsf{x}, t) = 0, \ \ \mathsf{x} \in \partial\Omega, \ \ \forall t. \tag{2.20}$$

The solution $c(\mathsf{x}, t)$ to (2.1) for a general source field $s(\mathsf{x}, t)$ can be obtained by superimposing the solutions (i.e., the Green's function) for unit impulses at various times, to yield:

$$c(\mathsf{x}, t) = \int_{-\infty}^{\infty} \int_{\Omega} g_{\mathsf{x}'t'}(\mathsf{x}, t) s(\mathsf{x}', t') \, d\mathsf{x}' dt'. \tag{2.21}$$

To derive this equation, just multiply both sides of (2.18) by $s(\mathsf{x}', t')$ and, then, integrate with respect to x' and t'. Note that the initial and boundary condition for the solution from (2.21) automatically follow from those placed upon $g_{\mathsf{x}'t'}(\mathsf{x}, t)$.

2.4. Reciprocal or Adjoint Green's Function

We have defined the (forward) Green's function $g_{\mathsf{x}'t'}(\mathsf{x}, t)$ as the solution to the equation (2.17). We now define the reciprocal or adjoint Green's function $h_{\mathsf{x}''t''}(\mathsf{x}, t)$ as the function, for $t < t''$, satisfies

$$\mathcal{L}^t[h_{\mathsf{x}''t''}(\mathsf{x}, t)] = \delta_{\mathsf{x}''t''}(\mathsf{x}, t), \tag{2.22}$$

and, for $t \geq t''$,

$$h_{\mathsf{x}''t''}(\mathsf{x}, t) = 0. \tag{2.23}$$

Here, $\mathcal{L}^t$ is the adjoint operator of $\mathcal{L}$, as defined by (2.16).

From our previous observations made regarding the adjoint operator, the reciprocal Green's function $h_{\mathsf{x}''t''}$ is the backward evolution of a concentration field due to a source at location x'' occurring at time t'' with time-reversed and direction reversed wind field (with proper time-reversal of λ and k that will described later).

2.5. Law of Reciprocity for Green's Functions

It can be shown (see Appendix B) that the following "reciprocity law" holds:

$$g_{\mathsf{x}'t'}(\mathsf{x}'', t'') = h_{\mathsf{x}''t''}(\mathsf{x}', t'). \tag{2.24}$$

The importance of this law for source inversion is the following: in order to determine the concentrations $g_{\mathsf{x}'t'}(\mathsf{x}'', t'')$ at, say, a monitoring point, x'' for a multitude of potential source locations x', we can instead solve for the concentration field $h_{\mathsf{x}''t''}(\mathsf{x}', t')$ at x' due to a source at the monitoring location x''. Since the number of monitoring locations is relatively small, it is much easier to compute $h_{\mathsf{x}''t''}(\mathsf{x}', t')$ than $g_{\mathsf{x}'t'}(\mathsf{x}'', t'')$.

In order to compute $h_{\mathsf{x}''t''}(\mathsf{x}', t')$ we need to solve (2.22). As we mentioned before, by reversing time, this equation is an advection-dispersion equation so that it can be solved using existing advection-dispersion computer codes with minor modifications, if any.

That reciprocity works for a *purely advective* system is not surprising because transport due to an incompressible flow field is clearly time-reversible. That is, symmetry (invariance) exists under time-reversal. For a combined advective-dispersive system, reciprocity is actually symmetry under time-reversal *plus* the interchange between source locations. From a purely physical point of view, this symmetry is not obvious. It turns out that reciprocity works because the dispersion term in the transport equation is self-adjoint. Self-adjointness is required (de Groot and Mazur, 1984) in order that the time rate of entropy for a closed system be non-negative. For statisticians, an equivalent statement is that the *information* content of a closed system can not increase.

2.5.1. Reciprocity for Steady Wind Field and Transport Parameters

Suppose that the wind field velocity and the transport parameters(i.e., decay constant λ and dispersion coefficient k) are constant in time:

$$\mathsf{v}(\mathsf{x},t) = \mathsf{v}(\mathsf{x}), \quad \lambda(\mathsf{x},t,\mathsf{v}(\mathsf{x},t)) = \lambda(\mathsf{x},\mathsf{v}(\mathsf{x})), \quad \mathsf{k}(\mathsf{x},t,\mathsf{v}(\mathsf{x},t)) = \mathsf{k}(\mathsf{x},\mathsf{v}(\mathsf{x})). \tag{2.25}$$

As shown in Appendix C, the above conditions imply the following.

- The Green's function $g_{\mathsf{x}'t'}(\mathsf{x},t)$ depends only the time difference $t-t'$ so that we may write $g_{\mathsf{x}'}(\mathsf{x},t-t')$ instead, or, simply, as $g_{\mathsf{x}'}(\mathsf{x},t)$
- The same holds for the reciprocal Green's function $h_{\mathsf{x}''t''}(\mathsf{x},t)$; we may write it as $h_{\mathsf{x}''}(\mathsf{x},t-t'')$, or, simply, as $h_{\mathsf{x}''}(\mathsf{x},t)$.
- Note that these Green's functions no longer depend on the time, t' or t'', of the respective source terms. Thus, we may choose a single time, say $t=0$, for the source term. Only a *single* Green's function need be computed, instead of one for each source term time.

Suppose that in addition to the conditions (2.25) we have that the transport parameters do not dependent on the sign of the wind field:

$$\lambda(\mathsf{x},\mathsf{v}(\mathsf{x})) = \lambda(\mathsf{x},-\mathsf{v}(\mathsf{x})), \quad \mathsf{k}(\mathsf{x},\mathsf{v}(\mathsf{x})) = \mathsf{k}(\mathsf{x},-\mathsf{v}(\mathsf{x})). \tag{2.26}$$

Then, a reciprocity law holds that is somewhat stronger than (2.24). In Appendix C we prove that

$$g_{\mathsf{x}'}(\mathsf{x}'',t) = \tilde{g}_{\mathsf{x}''}(\mathsf{x}',t), \tag{2.27}$$

where $\tilde{g}$ has the same definition as the Green's function g with the only difference being that the wind field direction is reversed (i.e., $\mathsf{v} = -\mathsf{v}$).

The identity (2.27) can be interpreted as the following: the concentration $c_1(t)$ at x'' due to a unit impulse source at $\mathsf{x} = \mathsf{x}'$, $t = 0$, carried by a wind field v is equal to the concentration $c_2(t)$ at the point x' due to a unit impulse source at $\mathsf{x} = \mathsf{x}''$, $t = 0$, carried by the *reverse* wind field $-\mathsf{v}$.

2.5.2. Reciprocity for Time-Varying Wind Field and Transport Parameters

Now we consider the case where the wind field v and the transport parameters λ and k depend on time. We show how the reciprocal Green's function $h_{\mathsf{x}''t''}(\mathsf{x}, t)$ can be computed using an existing computer code that solves advection-dispersion transport problems.

From (2.16) and (2.22), we see that the reciprocal Green's function solves the equation:

$$-\frac{\partial h_{\mathsf{x}''t''}(\mathsf{x},t)}{\partial t} + \lambda(\mathsf{x},t,\mathsf{v})h_{\mathsf{x}''t''}(\mathsf{x},t) - \nabla\cdot[h_{\mathsf{x}''t''}(\mathsf{x},t)\mathsf{v}(\mathsf{x},t)] \\ -\nabla\cdot\mathsf{k}(\mathsf{x},t,\mathsf{v})\nabla h_{\mathsf{x}''t''}(\mathsf{x},t) = \delta_{\mathsf{x}''t''}(\mathsf{x},t). \qquad (2.28)$$

We will use the variable τ to denote time that is reversed, starting from the release time t'' of the unit pulse source:

$$\tau = t'' - t. \qquad (2.29)$$

In this reversed time, we have that $\tau = 0$ corresponds to the release time $t = t''$. We define the reversed wind field as a reversal in time and direction:

$$\mathsf{v}^r_{t''}(\mathsf{x},\tau) \equiv -\mathsf{v}(\mathsf{x}, t'' - \tau). \qquad (2.30)$$

The reversed transport parameters are defined as the time reversal of the original parameters:

$$\lambda^r_{t''}(\mathsf{x},\tau,\mathsf{v}) \equiv \lambda(\mathsf{x}, t''-\tau, \mathsf{v}(\mathsf{x}, t''-\tau)), \qquad \mathsf{k}^r_{t''}(\mathsf{x},\tau,\mathsf{v}) \equiv \mathsf{k}(\mathsf{x}, t''-\tau, \mathsf{v}(\mathsf{x}, t''-\tau)). \qquad (2.31)$$

Note that, here, the wind velocity is not reversed.

The *reverse reciprocal Green's function* will be defined as simply the time-reversal of the reciprocal Green's function:

$$h^r_{\mathsf{x}''t''}(\mathsf{x},\tau) \equiv h_{\mathsf{x}''t''}(\mathsf{x}, t''-\tau). \qquad (2.32)$$

Then, (2.28) becomes, for $\tau > 0$,

$$\frac{\partial h^r_{\mathsf{x}''t''}(\mathsf{x},\tau)}{\partial\tau} + \lambda^r_{t''}(\mathsf{x},\tau,\mathsf{v})h^r_{\mathsf{x}''t''}(\mathsf{x},\tau) + \nabla\cdot[h^r_{\mathsf{x}''t''}(\mathsf{x},\tau)\mathsf{v}^r(\mathsf{x},\tau)] \\ -\nabla\cdot\mathsf{k}^r_{t''}(\mathsf{x},\tau,\mathsf{v})\nabla h^r_{\mathsf{x}''t''}(\mathsf{x},\tau) = \delta(\mathsf{x}-\mathsf{x}'')\delta(\tau), \qquad (2.33)$$

and (2.23) becomes, for $\tau \le 0$,

$$h^r_{\mathsf{x}''t''}(\mathsf{x},\tau) = 0. \tag{2.34}$$

These equations imply that the reversed reciprocal Green's function $h^r_{\mathsf{x}''t''}(\mathsf{x},\tau)$ is the concentration field due to an unit impulse source at $\tau = 0$ with wind field that is reversed in direction and time, and the transport parameters λ and k are reversed in time only. Hence, $h^r_{\mathsf{x}''t''}(\mathsf{x},\tau)$ may be computed using an existing advection-dispersion transport code with the proper changes in the wind field and transport parameters. Once $h^r_{\mathsf{x}''t''}(\mathsf{x},\tau)$ is computed, from (2.32) we may compute the reciprocal Green's function by

$$h_{\mathsf{x}''t''}(\mathsf{x},t) = h^r_{\mathsf{x}''t''}(\mathsf{x},t''-t). \tag{2.35}$$

In terms of the reversed reciprocal Green's function, the law of reciprocity (2.24) becomes

$$g_{\mathsf{x}'t'}(\mathsf{x}'',t'') = h^r_{\mathsf{x}''t''}(\mathsf{x}',t''-t'). \tag{2.36}$$

In terms of the source inversion problem, this identity may be interpreted as the following. To find the concentration $g_{\mathsf{x}'t'}(\mathsf{x}_m,t_m)$ at a monitoring location at $\mathsf{x} = \mathsf{x}_m$, $t = t_m$ due to a unit impulse source at $\mathsf{x} = \mathsf{x}'$, $t = t'$, one can equivalently solve the reverse problem (in the sense described above) for the concentration $h^r_{\mathsf{x}_m t_m}(\mathsf{x}',t_m - t')$ at $\mathsf{x} = \mathsf{x}'$, $t = t_m - t'$ due to a unit impulse source at $\mathsf{x} = \mathsf{x}_m$, $t = 0$.

2.6. Example and Verification of Reciprocity

We consider solving the one-dimensional advection-dispersion equation with a source at $x = x_s$. The concentration $c(x,t)$ solves

$$\frac{\partial c(x,t)}{\partial t} + v\frac{\partial c(x,t)}{\partial x} - \frac{\partial}{\partial x}k(x)\frac{\partial c(x,t)}{\partial x} = \delta(x-x_s), \quad x \in R^1. \tag{2.37}$$

The initial concentration is zero: $c(x,0) = 0$.

To verify reciprocity we will compare two solutions: the first solution $c_1(x,t)$ is due to a source at $x_s = 0$, and the second solution $c_2(x,t)$ is due to a source at $x_s = L$, where $L > 0$. In computing the second solution $c_2(x,t)$ we change the wind direction so that the velocity v is replaced by $-v$.

If reciprocity is true, then the concentration history $c_1(L,t)$ should be the same as the concentration history $c_2(0,t)$.

The velocity v is constant and uniform. The dispersion coefficient $k(x)$ is linear in x:

$$k(x) = \left(\frac{x}{L}\right)k_1 + \left(\frac{x-L}{L}\right)k_o, \quad x \in R^1, \tag{2.38}$$

such that $k(0) = k_o$ and $k(L) = k_1$. The Peclet number ($Pe \equiv vL/k$) for $k_o = 1.0$ is equal to 10.0, which implies that advection strongly dominates over dispersion in the domain around $x = 0$. The Peclet number corresponding to $k_1 = 10.0$ is 1.0 which means that dispersion and advection are about equal in their effect around $x = L$.

To find the approximate solutions $c_1(x,t)$ and $c_2(x,t)$, we use the particle method as implemented by an ensemble of `N` particles that executes a random walk:

$$x_{n+1} = x_n + \left[v + \frac{dk}{dx}(x_n)\right] \Delta t + \sqrt{2k(x_n)}\eta_n, \tag{2.39}$$

where η_n are independent samples from the gaussian distribution with zero mean and variance Δt. At initial time, $t = 0$, the particles begin at the source location, $x = x_s$ (which is $x_s = 0$ for the solution of c_1 and $x_s = L$ for the solution of c_2).

Table 2.1 shows the parameter values used in the simulations. The time step Δt size was chosen as

$$\Delta t = 0.2 \min \left\{\Delta x/v,\ (\Delta x)^2/k_{max}\right\}, \qquad k_{max} \equiv \max(k_0, k_1). \tag{2.40}$$

Figure 2.1 shows the concentration histories of the forward and reversed simulations, $c_1(x = L, t)$ and $c_2(x = 0, t)$, using 100,000 particles. There is good agreement between the two concentrations. Figure 2.2 shows the same concentration histories using a million particles. The agreement between the two concentrations becomes even closer, demonstrating the convergence of the particle algorithm to the same solution.

Figure 2.3 shows the concentration histories, except that the dk/dx term in (2.39) was removed in the algorithm. For this simulation, 100,000 particles were used. It can be shown from the theory of the Fokker-Planck equation (Reif, 1965; van Kampen, 1981) that without the dk/dx term the concentration $c(x,t)$ does not solve (2.37), but instead solves

$$\frac{\partial c(x,t)}{\partial t} + v\frac{\partial c(x,t)}{\partial x} - \frac{\partial^2}{\partial x^2}[k(x)c(x,t)] = \delta(x - x_s), \quad x \in R^1. \tag{2.41}$$

Since the dispersion term in this equation is not self-adjoint, the adjoint equation is no longer of the same form as the original equation with reversed wind direction. Thus, the two concentration histories c_1 and c_2 are not equal, as seen in Fig. 2.3. The dk/dx term is important in (2.39) when the magnitude of dk/dx becomes significant compared to the wind velocity.

3. Reciprocity for Atmospheric Transport Models with Memory

Lagrangian advection-dispersion transport models, such as that implemented in the LODI code used at NARAC, have dispersion that depends on the cumulative time from when each material packet is released at a source. The solutions to such models do not satisfy an advection-dispersion equation. However, the solutions can be mathematically

Parameter	Value
v	1.0
L	10.0
k_o	1.0
k_1	10.0
Δx	0.01

Table 2.1. Parameter Values Used in Particle Simulation

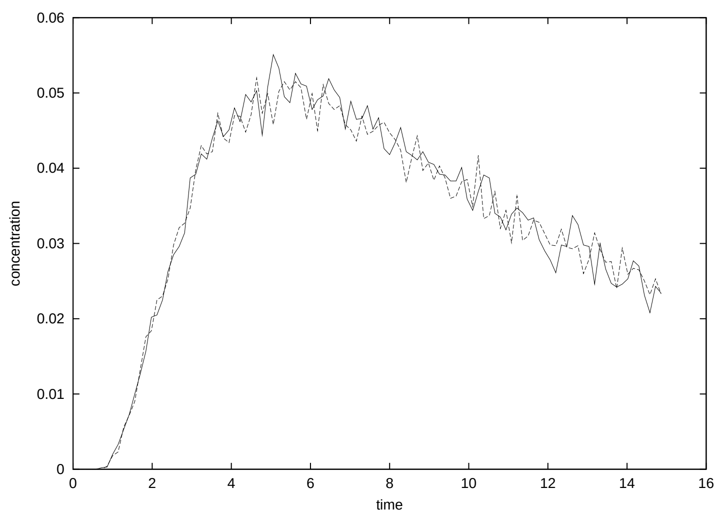

Figure 2.1. Concentration at $x = L$ for forward problem with source at $x = 0$ vs. concentration at $x = 0$ for the reverse problem with source at $x = L$. 100,000 particles were used.

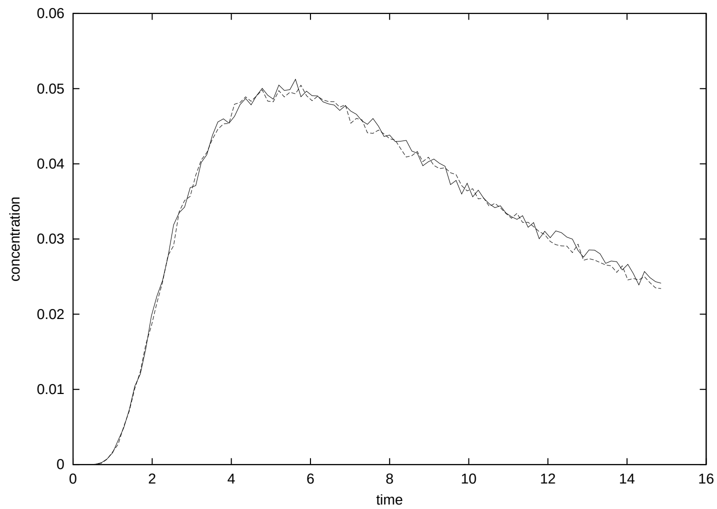

Figure 2.2. Same as Fig. 2.1 except that 1 million particles were used.

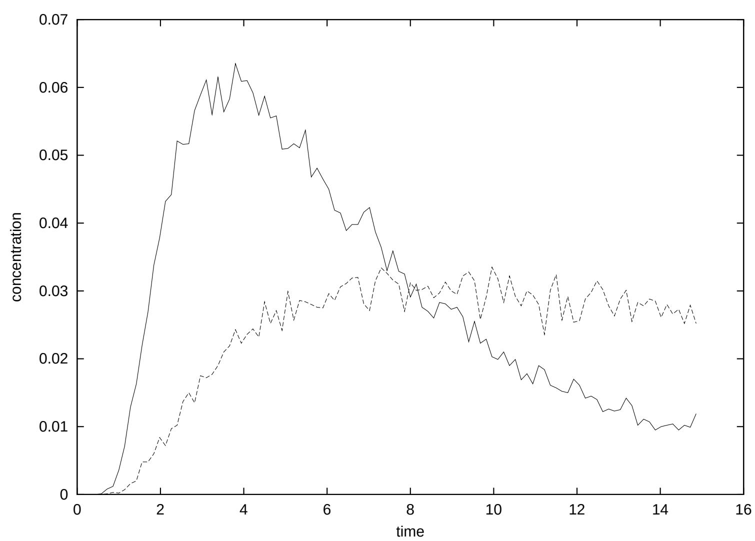

Figure 2.3. Same as Fig. 2.1 except that the dk/dx term in the random walk algorithm was neglected in both runs. 100,000 particles were used.

expressed as a superposition of point-source solutions each of whom satisfy a separate advection-dispersion equations that are parameterized with respect to the release time.

Each point source solution is a Green's function to an advection-dispersion equation for a specific release time. The reciprocal (adjoint) problem to LODI can still be formulated because the solution can be still written as a superposition of reciprocal (adjoint) Green's functions, using the law of reciprocity, for each specific release time.

When using the reciprocal Green's function approach for computing time-averaged concentrations, one has to be careful in computing time-averaged Green's functions from time-averaged point sources (i.e., square-wave). The time interval of averaging should not be larger than the time scale over which the dispersion coefficient varies. It may, perhaps, be better to first solve the problem for an instantaneous point source and then average the Green's function.

3.1. Reciprocal LODI Solutions

In general, we will consider releases from some time-varying source distribution $s(\mathsf{x}, t)$. As a special case, we shall also consider to a single point source located at a point x' with piecewise-constant $s(t'_j)$, $(j = 1, 2, ...)$, source strength history.

The mathematical model solved by LODI belongs to a class of Lagrangian advection-dispersion models that transport packets of material with an effective dispersion coefficient $\mathsf{k}(\mathsf{x}, t, t-t')$ that depends on the travel time $t-t'$ from when a packet was released. The solutions $c(\mathsf{x}, t)$ to such models do *not* solve an advection-dispersion equation of the form:

$$\frac{\partial c(\mathsf{x}, t)}{\partial t} + \nabla \cdot [\mathsf{v}(\mathsf{x}, t) c(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t) \nabla c(\mathsf{x}, t) = s(\mathsf{x}, t). \tag{3.1}$$

However, as shown in Appendix D, the solution can still be expressed in the form:

$$c(\mathsf{x}, t) = \int_0^t \int_\Omega g_{\mathsf{x}'t'}(\mathsf{x}, t) s(\mathsf{x}', t')\, d\mathsf{x}' dt', \tag{3.2}$$

where Ω stands for the spatial problem domain. For a piecewise-constant point source, a special case of (3.2) is

$$c(\mathsf{x}, t) = \sum_{j,\, t'_j < t} g_{\mathsf{x}'t'_j}(\mathsf{x}, t) s(t'_j) \Delta t'_j. \tag{3.3}$$

Each "basis function" $g_{\mathsf{x}'t'}(\mathsf{x}, t)$ is the concentration due to an instantaneous release at x', t' and solves an Eulerian advection-dispersion equation:

$$\frac{\partial g_{\mathsf{x}'t'}(\mathsf{x}, t)}{\partial t} + \nabla \cdot [g_{\mathsf{x}'t'}(\mathsf{x}, t) \mathsf{v}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, t-t') \nabla g_{\mathsf{x}'t'}(\mathsf{x}, t) = \delta(t-t')\delta(\mathsf{x}-\mathsf{x}'), \quad t > t', \tag{3.4}$$

where we note the presence of $t - t'$ in the argument of k, which distinguishes this equation from (3.1).

Note that $g_{\mathsf{x}'t'}(\mathsf{x}, t)$ is actually the Green's function of an advection-dispersion equation. But, the concentration $c(\mathsf{x}, t)$ will not, in general, solve an advection-dispersion equation because the equation solved by each Green's function has a distinct release time t'.

We can define an reciprocal, or adjoint, Green's function, $h_{\mathsf{x}''t''t'}(\mathsf{x}, t)$ so that the concentration may be re-expressed as

$$c(\mathsf{x}, t) = \int_0^t \int_\Omega h_{\mathsf{x}tt'}(\mathsf{x}', t') s(\mathsf{x}', t')\, d\mathsf{x}' dt'. \tag{3.5}$$

For a piecewise-constant point sources, this expression reduces to

$$c(\mathsf{x}, t) = \sum_{j,\, t'_j < t} h_{\mathsf{x}tt'_j}(\mathsf{x}', t'_j) s(t'_j) \Delta t'_j. \tag{3.6}$$

Here, $h_{\mathsf{x}''t''t'}(\mathsf{x}, t)$ solves the PDE:

$$-\frac{\partial h_{\mathsf{x}''t''t'}(\mathsf{x}, t)}{\partial t} - \nabla \cdot [\mathsf{v}(\mathsf{x}, t) h_{\mathsf{x}''t''t'}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, t - t') \nabla h_{\mathsf{x}''t''t'}(\mathsf{x}, t)$$
$$= \delta(t - t'') \delta(\mathsf{x} - \mathsf{x}''), \quad t' < t < t''. \tag{3.7}$$

Note that $h_{\mathsf{x}''t''t'}(\mathsf{x}, t)$ solves the adjoint to the problem in (3.4). Therefore, from the law of reciprocity, one has:

$$h_{\mathsf{x}''t''t'}(\mathsf{x}', t') = g_{\mathsf{x}'t'}(\mathsf{x}'', t''), \tag{3.8}$$

which, together with (3.2), implies (3.5).

We can also define the reverse reciprocal Green's function as

$$h^r_{\mathsf{x}''t''t'}(\mathsf{x}, \tau) \equiv h_{\mathsf{x}''t''t'}(\mathsf{x}, t'' - \tau), \tag{3.9}$$

where

$$\tau \equiv t'' - t. \tag{3.10}$$

It solves the equation

$$\frac{\partial h^r_{\mathsf{x}''t''t'}(\mathsf{x}, \tau)}{\partial \tau} + \nabla \cdot [h^r_{\mathsf{x}''t''t'}(\mathsf{x}, \tau) \mathsf{v}(\mathsf{x}, t'' - \tau)]$$
$$-\nabla \cdot \mathsf{k}(\mathsf{x}, t'' - \tau, t'' - \tau - t') \nabla h^r_{\mathsf{x}''t''t'}(\mathsf{x}, \tau) = \delta(\tau) \delta(\mathsf{x} - \mathsf{x}''),$$
$$0 < \tau \le t'' - t'. \tag{3.11}$$

The law of reciprocity implies

$$g_{\mathsf{x}'t'}(\mathsf{x}'',t'') = h^r_{\mathsf{x}''t''t'}(\mathsf{x}',t''-t'), \tag{3.12}$$

which can be used to express the concentrations in terms of h^r:

$$c(\mathsf{x},t) = \int_0^t \int_\Omega h^r_{\mathsf{x}tt'}(\mathsf{x}',t''-t')s(\mathsf{x}',t')\,d\mathsf{x}'dt'. \tag{3.13}$$

For a piecewise-constant point sources, this expression reduces to

$$c(\mathsf{x},t) = \sum_{j,\,t'_j<t} h^r_{\mathsf{x}tt'_j}(\mathsf{x}',t''-t'_j)s(t'_j)\Delta t'_j. \tag{3.14}$$

3.2. Computing Time-Averaged Reciprocal Green's Functions

The measurement of a concentration is usually the average over some time interval, say of length T:

$$\overline{c}(\mathsf{x},t) \equiv \frac{1}{T}\int_{t-\frac{1}{2}T}^{t+\frac{1}{2}T} c(\mathsf{x},t')\,dt'. \tag{3.15}$$

From (3.2), we have

$$\overline{c}(\mathsf{x},t) = \int_0^t \int_\Omega \overline{g}_{\mathsf{x}'t'}(\mathsf{x},t)s(\mathsf{x}',t')\,d\mathsf{x}'dt'. \tag{3.16}$$

where the time-averaged Green's function is defined by

$$\overline{g}_{\mathsf{x}'t'}(\mathsf{x},t) \equiv \frac{1}{T}\int_{t-\frac{1}{2}T}^{t+\frac{1}{2}T} g_{\mathsf{x}'t'}(\mathsf{x},\tilde{t})\,d\tilde{t}. \tag{3.17}$$

In order to compute $\overline{g}_{\mathsf{x}'t'}(\mathsf{x},t)$, one may be tempted to solve (3.4), except with a time-averaged unit point source:

$$\begin{aligned} s(\mathsf{x},t) &= \delta(\mathsf{x}-\mathsf{x}')\cdot\frac{1}{T}\int_{t-\frac{1}{2}T}^{t+\frac{1}{2}T}\delta(\tilde{t}-t')\,d\tilde{t} \\ &= \begin{cases} \frac{1}{T}\delta(\mathsf{x}-\mathsf{x}'), & t-\frac{1}{2}T < t < t+\frac{1}{2}T, \\ 0, & \text{otherwise.} \end{cases} \end{aligned} \tag{3.18}$$

From examining (3.4), it is seen that the resulting solution is not equal to $\overline{g}_{\mathsf{x}'t'}(\mathsf{x},t)$ even if the velocity is constant in time, due to the dependence on $t-t'$ by k. It will, however, be approximately equal to $\overline{g}_{\mathsf{x}'t'}(\mathsf{x},t)$ if v and k vary slowly over the relevant time intervals of length T. It may be possible that this condition is not as severe if, instead, we solve (3.4) and then compute the average according to (3.17).

Of course, the discussion in this section also applies to the computation of the reciprocal Green's function h and the reverse reciprocal Green's function h^r.

4. Reciprocal Solutions to "Puff Models"

A "puff model" solves the atmospheric transport of material in an approximate manner by treating the plume as a linear superposition of "puff functions" released at certain time interfvals from a source. These functions are typically gaussians with means translated by the velocity field and whose variance increases with time due to dispersive mixing. The concentration field computed by puff models, doe not, in general, satisfy the advection diffusion partial differential equation. However, in some cases, each puff function satisfies such an equation or approximately satisfies it.

In this section we show how to implement reciprocity for the puff model in the well-known code INPUFF (Petersen et al., 1984). Instead of an adjoint Green's function, we define a related concept, the "adjoint, or reciprocal, puff function". We show that for INPUFF, this function can be computed using the code as a "black-box" (i.e., without any modification to it), as long as the wind field does not vary significantly over a length scale on the order of the maximum puff width. No other assumptions are required. The black-box approach requires a backward traverse to compute the puff path trajectory, followed by a forward traverse to compute the dispersive standard deviation σ.

4.1. Adjoint Puff-Function

The concentration $c(x, y, z, t)$ computed from a puff model is a superposition of time-shifted "puff-functions":

$$c(x, y, z, t) = \sum_j g_{x_s y_s z_s t_{s,j}}(x, y, z, t) s(t_{s,j}) \Delta t_j, \tag{4.1}$$

where $t_{s,j}$ is the time of the j-th packet release (with $t_{s,j} < t$) and (x_s, y_s, z_s) is the coordinate of the (point) source location. Suppose that we measure concentration at a monitoring point (x_m, y_m, z_m) so that we have

$$c(x_m, y_m, z_m, t) = \sum_j g_{x_s y_s z_s t_{s,j}}(x_m, y_m, z_m, t) s(t_{s,j}) \Delta t_j. \tag{4.2}$$

We would like to find adjoint functions $h_{x_m y_m z_m t}$ such that

$$h_{x_m y_m z_m t}(x_s, y_s, z_s, t_s) = g_{x_s y_s z_s t_s}(x_m, y_m, z_m, t). \tag{4.3}$$

Then, we would have

$$c(x_m, y_m, z_m, t) = \sum_j h_{x_m y_m z_m t}(x_s, y_s, z_s, t_{s,j}) s(t_{s,j}) \Delta t_j. \tag{4.4}$$

In this way, we would only need to compute the function $h_{x_m y_m z_m t}(x, y, z, t_s)$, once for each t_s instead of having to compute $g_{x_s y_s z_s t_s}(x_m, y_m, z_m, t)$ for each potential source location (x_s, y_s, z_s). We will call the function h the "adjoint puff-function".

4.2. INPUFF Puff-Function

Suppose that a puff leaves the point source (x_s, y_s, z_s) at time t_s and is located at the point $(x_p(t), y_p(t), z_p(t))$ at time t. In INPUFF, the puff-function is of the form:

$$g_{x_s y_s z_s t_s}(x,y,z,t) =$$
$$\frac{1}{(2\pi)^{3/2}\sigma_r^2\big|_t \sigma_z\big|_t} \exp\left[-\frac{1}{2}\left(\frac{x - x_p\big|_t}{\sigma_r\big|_t}\right)^2\right] \exp\left[-\frac{1}{2}\left(\frac{y - y_p\big|_t}{\sigma_r\big|_t}\right)^2\right]$$
$$\cdot\left\{\exp\left[-\frac{1}{2}\left(\frac{z + z_p\big|_t}{\sigma_z\big|_t}\right)^2\right] + \exp\left[-\frac{1}{2}\left(\frac{z - z_p\big|_t}{\sigma_z\big|_t}\right)^2\right]\right\}. \quad (4.5)$$

The vertical puff position is given by the "plume rise" formulation (call to subroutine `PLUMRS`) which has the functional form:

$$z_p\Big|_t = z_p(k_s(t), w_s(x_p, y_p, t), T(t), H_s), \quad (4.6)$$

$k_s(t)$	(`KS`) stability class (from met. data records),
$w_s = w_s(t)$	(`WSPD`)wind speed (from met. data records),
$w_s = w_s(x_p, y_p, t)$	(`GRIDU`, `GRIDV`) wind speed on a grid (from user met. file),
$T(t)$	(`TEMP`) temperature (from met. data records),
H_s	(`HPP`) vertical height of release point (read from input, not the same as $z_p(t_s)$).

The horizontal puff velocity vector (u_p, v_p) is given by

$$u_p(x_p, y_p, z_p, t) = u_w(x, y, t) \cdot \alpha(z_p, k_s(t)), \quad (4.7)$$

$$v_p(x_p, y_p, z_p, t) = v_w(x, y, t) \cdot \alpha(z_p, k_s(t)), \quad (4.8)$$

with the wind velocity given by

$$u_w(x,y,t) = w_s(t)\cos\theta_w(t)\pi/180, \qquad v_w(x,y,t) = w_s(t)\sin\theta_w(t)\pi/180, \quad (4.9)$$

where $w_s(t)$ (`WSPD`) and $\theta_w(t)$ (`WDIR`) are from the met. data records. Another option is

$$u_w(x,y,t) = w_s(x,y,t)\cos\theta_w(x,y,t)\pi/180, \quad v_w(x,y,t) = w_s(x,y,t)\sin\theta_w(x,y,t)\pi/180, \quad (4.10)$$

with $w_s(x,y,t)$ and $\theta_w(x,y,t)$ from the user met. file (`GRIDU` and `GRIDV`). Here, $\alpha(z_p, k_s(t))$ (`USCAL`) is a plume height-dependent factor. The path of the puff on the horizontal x-y plane obeys the ordinary differential equations:

$$\frac{dx_p}{dt} = u_p(x_p, y_p, z_p\big|_t, t), \quad \frac{dy_p}{dt} = v_p(x_p, y_p, z_p\big|_t, t), \quad (4.11)$$

with initial conditions:

$$x_p\Big|_{t=t_s} = x_s, \qquad y_p\Big|_{t=t_s} = y_s. \tag{4.12}$$

The formulation for computing σ_r and σ_z (called `SY` and `SZ`; computed in subroutine `PROCES`) can be set to one of two possible functional forms denpending on a flag (`KEYDSP`) specified by the user. The first form is based on the time travelled $t - t_s$ by the puff:

$$\sigma_r\Big|_t = \sigma_r(t - t_s, k_s(t), \lambda_v(x, y, t)), \tag{4.13}$$

$$\sigma_z\Big|_t = \sigma_r(t - t_s, k_s(t), \lambda_w(x, y, t), H_{mix}(t)). \tag{4.14}$$

The second is based on the distance travelled by the puff from the source:

$$\sigma_r\Big|_t = \sigma_r(s\Big|_t, k_s(t), \lambda_v(x, y, t)), \tag{4.15}$$

$$\sigma_z\Big|_t = \sigma_z(s\Big|_t, k_s(t), \lambda_w(x, y, t), H_{mix}(t)). \tag{4.16}$$

The travel distance $s\Big|_t$ is computed by integrating the differential equation:

$$\frac{ds}{dt} = \sqrt{u_p^2\Big|_t + v_p^2\Big|_t}, \qquad s\Big|_{t=t_s} = 0. \tag{4.17}$$

Here,

$$\lambda_v(x, y, t) = u_w(x, y, t)\sigma_v(t), \qquad \lambda_w(x, y, t) = v_w(x, y, t)\sigma_w(t), \tag{4.18}$$

where $\sigma_v(t)$ and $\sigma_w(t)$ are standard deviations of uncertainty in the wind (read from the met. data records). [**Note:** There appears to be an error in the code in the above computation when velocities are read from a met. file. Subroutine `PROCESS` uses the variables `SV` and `SW` for λ_v and λ_w which are computed in `CMPRIS` based on the met. record wind velocity, but they should instead be computed from the met. file wind velocity.] The quantity $H_{mix}(t)$ is the thickness of the "mixed depth layer" (`HL`).

When $\sigma_z\Big|_t > 0.8H_{mix}(t)$, equation (4.5) is replaced with

$$g_{x_s y_s z_s t_s}(x, y, z, t) = \frac{1}{2\pi\sigma_r^2\Big|_t H_{mix}(t)} \exp\left[-\frac{1}{2}\left(\frac{x - x_p\Big|_t}{\sigma_r\Big|_t}\right)^2\right] \exp\left[-\frac{1}{2}\left(\frac{y - y_p\Big|_t}{\sigma_r\Big|_t}\right)^2\right]. \tag{4.19}$$

4.3. Computing the Adjoint, or Reciprocal, Puff-Function

We now consider the adjoint, or reciprocal, puff function for INPUFF. Actually, we shall not exhibit the adjoint puff function directly but we shall show how to computed it indirectly using the code as a "black-box".

First, we observe that the following assumption (Assumption A.) must hold for the INPUFF puff model to be physically correct: the spatial variation in the wind velocity vector is small over length scales equal to the maximum width of the puff:

$$|\mathsf{v}_w(\mathsf{r}_1, t) - \mathsf{v}_w(\mathsf{r}_2, t)| \cdot t \ll \sigma_r(t), \tag{4.20}$$

for

$$|\mathsf{r}_1 - \mathsf{r}_2| < \eta(\sigma_r)_{max}, \tag{4.21}$$

where η is sufficiently large enough that the error in computing the puff-function is small compared to the smallest meaningful magnitude in concentration for the problem. Here, v_w is the wind field velocity vector.

We make the following observations regarding the INPUFF puff function:

Observation 1. The computation of x_p, y_p, z_p is independent of σ_r and σ_z.

Observation 2. The puff height z_p is purely a function of t, x_p, y_p. Therefore, the differential equation (4.11) is of the form:

$$\frac{dx_p}{dt} = f_x(x_p, y_p, t), \qquad \frac{dy_p}{dt} = f_y(x_p, y_p, t). \tag{4.22}$$

Observation 3. If we reverse the wind field and integrate backwards in time from $t = t_m$ to $t = t_s$, starting at the monitoring point M (see Fig. 4.1), we end up at a point S* which is approximately the same distance as from M to the true puff position P. This observation follows from Assumption A, above.

From these observations, one can compute $g_{x_s y_s z_s t_s}(x, y, z, t)$ at x_m, y_m, z_m, t_m using the following steps:

Step 1. One reverses the wind field and shifts it in time so that $t = t_m$ becomes $t = 0$. One then calls INPUFF with the source located at point M (see Fig. 4.1). A single puff is released at time $t = 0$. The stopping time is at $t = t_m - t_s$. The puff coordinate position (point S* in Fig. 1) is output at this time. In effect, we are solving (4.22) backwards in time from M in order to obtain the coordinate of S*.

Step 2. One then calls INPUFF again; this time using the original wind field. The source is located at point S*. A single puff is released at $t = t_s$ and the run stops at $t = t_m$. In effect, we are solving (4.22) forward in time from t_s to t_m starting from S* using the original wind field. We do this in order to compute the correct values for σ_r and σ_z. The end of the path ends at some point M* (not pictured) that is close to M, but not exactly, due to numerical errors in computing the backward and forward paths. Suppose we call the resulting puff function centered at M* by $\psi_{SM}(x, y, z; t_s, t_m)$.

Step 3. At the end of INPUFF run described in Step 2., the concentration computed by the code at the point P* (shown in Fig. 4.1) at time t_m is approximately equal to the desired concentration due to a puff centered at P starting from x_s. That is,

$$g_{x_s y_s z_s t_s}(x_m, y_m, z_m, t_m) = \psi_{SM}(x_{P^*}, y_{P^*}, z_{P^*}; t_s, t_m). \tag{4.23}$$

(Actually, instead of using M to compute the coordinates of P*, as shown in the figure, we should, instead, use M* computed from $\mathsf{r}_{P^*} = \mathsf{r}_{M^*} + \mathsf{r}_1$ where $\mathsf{r}_1 = \overrightarrow{\mathrm{S^*S}}$.)

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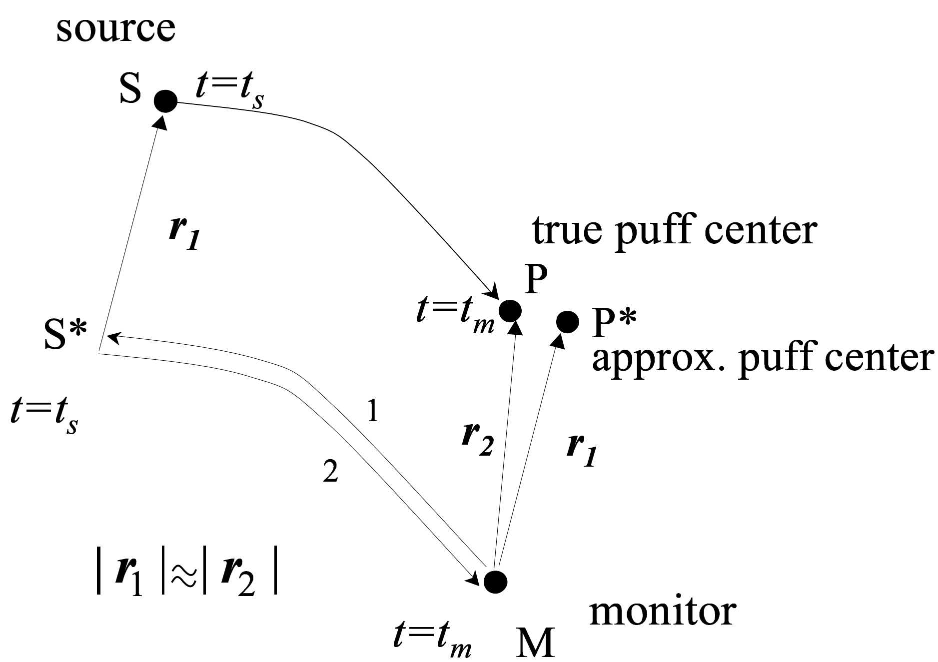

Figure 4.1. First, INPUFF is run from $t = t_m$ to t_s starting at M in the reversed wind field. Then, INPUFF is run from $t = t_s$ to t_m starting from S*. The puff evaluated at $t = t_m$ at P* is approximately the same as the desired concentration one would get from the puff at P. In fact, it is exactly the same if the wind field is such that the streak-lines from S to P and S* to M are parallel, ignoring any numerical errors in path integration.

A. Appendix: Derivation of the Adjoint to the Advection-Dispersion Equation

We define the space-time inner product $< . , . >$ by

$$< \phi, \psi > \equiv \int_{-\infty}^{\infty} \int_{\Omega} \phi(\mathsf{x}, t)\psi(\mathsf{x}, t)\, d\mathsf{x}dt. \tag{A.1}$$

In this section, we only consider functions $\phi(\mathsf{x})$ that satisfy the following conditions:

$$\lim_{t\to\infty} \int_{\Omega} \phi(\mathsf{x}, t)^2\, d\mathsf{x} = \lim_{t\to-\infty} \int_{\Omega} \phi(\mathsf{x}, t)^2\, d\mathsf{x} = 0, \tag{A.2}$$

$$\phi(\mathsf{x}, t) = 0,\ \mathsf{x} \in \partial\Omega,\ \forall t. \tag{A.3}$$

Operator	Adjoint	Corresponding Identity
$\phi \to f\phi$	$\psi \to f\psi$	(I1) $< \psi, f\phi > = < f\psi, \phi >$
$\phi \to \partial\phi/\partial t$	$\psi \to -\partial\psi/\partial t$	(I2) $< \psi, \partial\phi/\partial t >= < -\partial\psi/\partial t, \phi >$
$\mathsf{u} \to \nabla \cdot \mathsf{u}$	$\psi \to -\nabla\psi$	(I3) $< \psi, \nabla \cdot \mathsf{u} > = < -\nabla\psi, \mathsf{u} >$
$\phi \to \nabla\phi$	$\mathsf{u} \to -\nabla \cdot \mathsf{u}$	(I4) $< \mathsf{u}, \nabla\phi > = < -\nabla \cdot \mathsf{u}, \phi >$

Table A.1. Relevant operators and their adjoints.

For vector-valued functions $\mathsf{u}(\mathsf{x}, t)$ and $\mathsf{w}(\mathsf{x}, t)$ we define the inner product by

$$< \mathsf{u}, \mathsf{w} >\equiv \int_{-\infty}^{\infty} \int_{\Omega} \mathsf{u}(\mathsf{x}, t) \cdot \mathsf{w}(\mathsf{x}, t)\, d\mathsf{x}dt. \tag{A.4}$$

However, we do *not* require the vector-valued functions to satisfy conditions of the type (A.2) and (A.3).

Given a linear operator $\mathcal{O}$ its adjoint $\mathcal{O}^t$ is defined to be the operator such that

$$< \mathcal{O}\phi, \psi >=< \phi, \mathcal{O}^t\psi > . \tag{A.5}$$

Or, for an operator $\mathcal{A}$ that maps real-valued to vector-valued functions, its adjoint $\mathcal{A}^t$ must map vector-valued to real-valued functions and is defined such that it satisfies

$$< \mathcal{A}\phi, \mathsf{u} >=< \phi, \mathcal{A}^t\mathsf{u} > . \tag{A.6}$$

For an operator $\mathcal{B}$ that maps vector-valued to real-valued functions, its adjoint $\mathcal{B}^t$ must map real-valued to vector-valued functions and must satisfy

$$< \mathcal{B}\mathsf{u}, \phi >=< \mathsf{u}, \mathcal{B}^t\phi > . \tag{A.7}$$

Table A.1 shows some relevant operators and their adjoints. Also, shown are the corresponding identities from which the adjoint is derived. We will now prove the identities in this table.

The first identity (I1) in the Table is obvious. The second identity (I2) follows from performing the following integration by parts:

$$\begin{aligned} < \psi, \frac{\partial\phi}{\partial t} > &= \int_{-\infty}^{\infty} \int_{\Omega} \psi \frac{\partial\phi}{\partial t}\, d\mathsf{x}dt \\ &= \int_{-\infty}^{\infty} \int_{\Omega} \frac{\partial(\psi\phi)}{\partial t}\, d\mathsf{x}dt - \int_{-\infty}^{\infty} \int_{\Omega} \frac{\partial\psi}{\partial t}\phi\, d\mathsf{x}dt \\ &= \left[\int_{\Omega} \psi\phi\, d\mathsf{x}\right]_{t=-\infty}^{t=\infty} - \int_{-\infty}^{\infty} \int_{\Omega} \frac{\partial\psi}{\partial t}\phi\, d\mathsf{x}dt. \end{aligned} \tag{A.8}$$

The magnitude of the first integral on the last r.h.s. equation is bounded above by the Cauchy-Schwarz inequality:

$$\left|\int_\Omega \psi\phi\, d\mathsf{x}\right| \leq \left(\int_\Omega \psi^2\, d\mathsf{x} \int_\Omega \phi^2\, d\mathsf{x}\right)^{1/2}. \tag{A.9}$$

From (A.2), the two integrals on the r.h.s. of the inequality both go to zero as $t- \to \infty$ and $t \to \infty$. Hence, the integral on the r.h.s. of (A.8) vanishes and we have

$$< \psi, \frac{\partial \phi}{\partial t} >= -\int_{-\infty}^{\infty}\int_\Omega \frac{\partial \psi}{\partial t}\phi\, d\mathsf{x}dt =< -\frac{\partial \psi}{\partial t}, \phi > . \tag{A.10}$$

To prove identity (I3) in Table A.1, we have

$$\begin{aligned} < \psi, \nabla \cdot \mathsf{u} > &= \int_{-\infty}^{\infty}\int_\Omega \psi \nabla \cdot \mathsf{u}\, d\mathsf{x}dt \\ &= \int_{-\infty}^{\infty}\int_\Omega \nabla \cdot (\psi \mathsf{u})\, d\mathsf{x}dt - \int_{-\infty}^{\infty}\int_\Omega \mathsf{u} \cdot \nabla\psi\, d\mathsf{x}dt. \end{aligned} \tag{A.11}$$

From the divergence theorem, the above equation becomes

$$< \psi, \nabla \cdot \mathsf{u} > = \int_{-\infty}^{\infty}\int_{\partial\Omega} (\psi \mathsf{u}) \cdot \mathsf{n}\, dSdt - \int_{-\infty}^{\infty}\int_\Omega \mathsf{u} \cdot \nabla\psi\, d\mathsf{x}dt. \tag{A.12}$$

From condition (A.3) we have that ψ vanishes on the boundary $\partial\Omega$ so that the first integral on the r.h.s. vanishes, and identity (I3) immediately follows.

Identity (I4) immediately follows from (I3) and using the commutativity of the inner product $< . , . >$.

The operator for advection-dispersion is

$$\mathcal{L}[c(\mathsf{x}, t)] \equiv \frac{\partial c(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, t, \mathsf{v})c(\mathsf{x}, t) + \nabla \cdot [c(\mathsf{x}, t)\mathsf{v}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, \mathsf{v})\nabla c(\mathsf{x}, t). \tag{A.13}$$

We shall assume that the flow is incompressible; that is,

$$\nabla \cdot \mathsf{v}(\mathsf{x}, t) = 0. \tag{A.14}$$

To find the adjoint of $\mathcal{L}$, we take the adjoint of each of the above terms in the r.h.s. of (A.13) by using primarily the results in Table A.1.

In particular, from (I2) the first term on the r.h.s. (A.13), upon taking its adjoint, becomes $-\partial c(\mathsf{x}, t)/\partial t$.

From (I1), the adjoint of the second term is $\lambda(\mathsf{x}, t)c(\mathsf{x}, t)$, which is left unchanged.

The adjoint of the third term (advective term) in (A.13) is derived using the incompressibility assumption (A.14) and applying (I4). We have

$$
\begin{aligned}
<\psi, \nabla\cdot(\phi \mathsf{v})> &= <\psi, \mathsf{v}\cdot\nabla\phi> + <\psi, \phi\nabla\cdot\mathsf{v}> \\
&= <\psi, \mathsf{v}\cdot\nabla\phi> \\
&= <\psi\mathsf{v}, \nabla\phi> \\
&= <-\nabla\cdot(\psi\mathsf{v}), \phi> . \qquad \text{(A.15)}
\end{aligned}
$$

Thus, the adjoint of the advection operator is its negation.

The adjoint of the last term (dispersion term), is obtained by the following identity:

$$
\begin{aligned}
<-\psi, \nabla\cdot\mathsf{k}\nabla\phi> &= <\nabla\psi, \mathsf{k}\nabla\phi> \\
&= <\mathsf{k}\nabla\psi, \nabla\phi> \\
&= <-\nabla\cdot\mathsf{k}\nabla\psi, \phi>, \qquad \text{(A.16)}
\end{aligned}
$$

where we used the fact that the dispersion tensor k is symmetric. Thus, the adjoint of the dispersion operator is itself; i.e., it is self-adjoint.

Thus, the adjoint of $\mathcal{L}$ is finally given by

$$
\mathcal{L}^t[c(\mathsf{x},t)] \equiv -\frac{\partial c(\mathsf{x},t)}{\partial t} + \lambda(\mathsf{x},t,\mathsf{v})c(\mathsf{x},t) - \nabla\cdot[c(\mathsf{x},t)\mathsf{v}(\mathsf{x},t)] - \nabla\cdot\mathsf{k}(\mathsf{x},t,\mathsf{v})\nabla c(\mathsf{x},t). \quad \text{(A.17)}
$$

B. Appendix: Derivation of the Reciprocity Relationship for the Green's Function

The Green's function $g_{\mathsf{x}'t'}(\mathsf{x},t)$ satisfies the problem:

$$
\mathcal{L}\, g_{\mathsf{x}'t'}(\mathsf{x},t) = \delta_{\mathsf{x}'t'}(\mathsf{x},t). \qquad \text{(B.1)}
$$

Here, $\delta_{\mathsf{x}'t'}(\mathsf{x},t)$ denotes the delta function centered at $x = \mathsf{x}'$ occurring at time $t = t'$. The adjoint, or reciprocal, Green's function $h_{\mathsf{x}''t''}(\mathsf{x},t)$ satisfies the adjoint problem:

$$
\mathcal{L}^t\, h_{\mathsf{x}''t''}(\mathsf{x},t) = \delta_{\mathsf{x}''t''}(\mathsf{x},t). \qquad \text{(B.2)}
$$

Using the inner product defined in (A.1), it immediately follows that

$$
\begin{aligned}
<h_{\mathsf{x}''t''}, \mathcal{L}g_{\mathsf{x}'t'}> &= <h_{\mathsf{x}''t''}, \delta_{\mathsf{x}'t'}> \\
&= \int_{-\infty}^{\infty}\int_{\Omega} h_{\mathsf{x}''t''}(\mathsf{x},t)\delta_{\mathsf{x}'t'}(\mathsf{x},t)\,\mathsf{d}\mathsf{x}dt \\
&= h_{\mathsf{x}''t''}(\mathsf{x}',t'). \qquad \text{(B.3)}
\end{aligned}
$$

From the definition of the adjoint operator, the above inner product can also be expressed as

$$
\begin{aligned}
< h_{\mathsf{x}''t''}, \mathcal{L} g_{\mathsf{x}'t'} > &= < \mathcal{L}^t h_{\mathsf{x}''t''}, g_{\mathsf{x}'t'} > \\
&= < \delta_{\mathsf{x}''t''}, g_{\mathsf{x}'t'} > \\
&= \int_{-\infty}^{\infty} \int_{\Omega} \delta_{\mathsf{x}''t''}(\mathsf{x}, t) g_{\mathsf{x}'t'}(\mathsf{x}, t)\, \mathsf{d}\mathsf{x} dt \\
&= g_{\mathsf{x}'t'}(\mathsf{x}'', t''). \qquad \text{(B.4)}
\end{aligned}
$$

Thus, by equating the last lines of (B.3) and (B.4), we have the law of reciprocity:

$$
g_{\mathsf{x}'t'}(\mathsf{x}'', t'') = h_{\mathsf{x}''t''}(\mathsf{x}', t'). \qquad \text{(B.5)}
$$

C. Appendix: Derivation of the "Strong" Reciprocity Law in the Case of Steady Wind Field and Transport Parameters

We assume that the wind field velocity, decay constant, and dispersion coefficient are constant in time. That is,

$$
\mathsf{v}(\mathsf{x}, t) = \mathsf{v}(\mathsf{x}), \qquad \lambda(\mathsf{x}, t, \mathsf{v}(\mathsf{x}, t)) = \lambda(\mathsf{x}, \mathsf{v}(\mathsf{x})), \qquad \mathsf{k}(\mathsf{x}, t, \mathsf{v}(\mathsf{x}, t)) = \mathsf{k}(\mathsf{x}, \mathsf{v}(\mathsf{x})). \qquad \text{(C.1)}
$$

Moreover, we assume that the decay and dispersion coefficients do not depend on the sign of the wind velocity. That is,

$$
\lambda(\mathsf{x}, \mathsf{v}) = \lambda(\mathsf{x}, -\mathsf{v}), \qquad \mathsf{k}(\mathsf{x}, \mathsf{v}) = \mathsf{k}(\mathsf{x}, -\mathsf{v}). \qquad \text{(C.2)}
$$

Under these assumptions, the equation (2.18) defining the (forward) Green's function becomes

$$
\begin{aligned}
\frac{\partial g_{\mathsf{x}'t'}(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, \mathsf{v}) g_{\mathsf{x}'t'}(\mathsf{x}, t) + \nabla \cdot [g_{\mathsf{x}'t'}(\mathsf{x}, t)\mathsf{v}(\mathsf{x})] \\
-\nabla \cdot \mathsf{k}(\mathsf{x}, \mathsf{v}) \nabla g_{\mathsf{x}'t'}(\mathsf{x}, t) = \delta_{\mathsf{x}'t'}(\mathsf{x}, t). \qquad \text{(C.3)}
\end{aligned}
$$

Because the coefficients λ, v, and k in the equation are not functions of t, it can be easily shown that if $g_{\mathsf{x}'t'}(\mathsf{x}, t)$ is a solution to this equation, then $g_{\mathsf{x}'t'+\tau}(\mathsf{x}, t+\tau)$ is also a solution, for any constant τ. Thus, the Green's function, in this case, only depends on the difference $t - t'$. Thus, we may simply express the Green's function by $g_{\mathsf{x}'}(\mathsf{x}, t - t')$, or simply $g_{\mathsf{x}'}(\mathsf{x}, t)$, instead of $g_{\mathsf{x}'t'}(\mathsf{x}, t)$. Equation (C.3) becomes, for $t > 0$,

$$
\begin{aligned}
\frac{\partial g_{\mathsf{x}'}(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, \mathsf{v}) g_{\mathsf{x}'}(\mathsf{x}, t) + \nabla \cdot [g_{\mathsf{x}'}(\mathsf{x}, t)\mathsf{v}(\mathsf{x})] \\
-\nabla \cdot \mathsf{k}(\mathsf{x}, \mathsf{v}) \nabla g_{\mathsf{x}'}(\mathsf{x}, t) = \delta(\mathsf{x} - \mathsf{x}')\delta(t), \qquad \text{(C.4)}
\end{aligned}
$$

and condition (2.23) becomes, for $t \leq 0$,

$$g_{\mathsf{x}'}(\mathsf{x}, t) = 0. \tag{C.5}$$

Note that t' no longer appears in either of these equations.

By a similar argument, the reciprocal Green's function only depends on the difference $t - t''$ so that we may write it as $h_{\mathsf{x}''}(\mathsf{x}, t - t'')$, or simply $h_{\mathsf{x}''}(\mathsf{x}, t)$. Equation (2.22) for the reciprocal Green's function becomes, for $t < 0$,

$$\begin{aligned} -\frac{\partial h_{\mathsf{x}''}(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, \mathsf{v}) h_{\mathsf{x}''}(\mathsf{x}, t) - \nabla \cdot [h_{\mathsf{x}''}(\mathsf{x}, t)\mathsf{v}(\mathsf{x})] \\ -\nabla \cdot \mathsf{k}(\mathsf{x}, \mathsf{v}) \nabla h_{\mathsf{x}''}(\mathsf{x}, t) = \delta(\mathsf{x} - \mathsf{x}'')\delta(t), \end{aligned} \tag{C.6}$$

and, for $t \geq 0$,

$$h_{\mathsf{x}'}(\mathsf{x}, t) = 0. \tag{C.7}$$

Note that t'' no longer appears in either of these equations.

In equation (C.6), let us reverse the time variable, that is, we make the substitution:

$$t \to -t. \tag{C.8}$$

The time-reversed reciprocal Green's function is defined by

$$h^r_{\mathsf{x}''}(\mathsf{x}, t) \equiv h_{\mathsf{x}''}(\mathsf{x}, -t). \tag{C.9}$$

We also define the reversed wind field by

$$\mathsf{v}^r(\mathsf{x}) = -\mathsf{v}(\mathsf{x}). \tag{C.10}$$

Then, making use of (C.2), equation (C.6) becomes, for $t > 0$,

$$\begin{aligned} \frac{\partial h^r_{\mathsf{x}''}(\mathsf{x}, t)}{\partial t} + \lambda(\mathsf{x}, \mathsf{v}^r) h^r_{\mathsf{x}''}(\mathsf{x}, t) + \nabla \cdot [h^r_{\mathsf{x}''}(\mathsf{x}, t)\mathsf{v}^r(\mathsf{x})] \\ -\nabla \cdot \mathsf{k}(\mathsf{x}, \mathsf{v}^r) \nabla h^r_{\mathsf{x}''}(\mathsf{x}, t) = \delta(\mathsf{x} - \mathsf{x}'')\delta(t), \end{aligned} \tag{C.11}$$

and condition (2.23) becomes, for $t \leq 0$,

$$h^r_{\mathsf{x}''}(\mathsf{x}, t) = 0. \tag{C.12}$$

It is important to note that, except for the reversal in wind field, equation (C.11) for the reverse reciprocal Green's function is exactly of the same form as (C.4) for the forward Green's function. Thus, $h^r_{\mathsf{x}'}(\mathsf{x}, t)$ is a "reversed wind-field" Green's function corresponding to $g_{\mathsf{x}'}(\mathsf{x}, t)$. Note that in the main text we used the notation $\tilde{g}$ instead of h^r.

From the reciprocity law given by (2.24), we note that

$$g_{\mathsf{x}'}(\mathsf{x}'', t'' - t') = g_{\mathsf{x}'t'}(\mathsf{x}'', t'') = h_{\mathsf{x}''t''}(\mathsf{x}', t') = h_{\mathsf{x}''}(\mathsf{x}', t' - t'') = h^r_{\mathsf{x}''}(\mathsf{x}', t'' - t'). \quad \text{(C.13)}$$

Setting $t = t'' - t'$, we then have the reciprocity law for the reversed reciprocal Green's function:

$$g_{\mathsf{x}'}(\mathsf{x}'', t) = h^r_{\mathsf{x}''}(\mathsf{x}', t). \quad \text{(C.14)}$$

This identity states that the concentration $c_1(t) = g_{\mathsf{x}'}(\mathsf{x}'', t)$ at $\mathsf{x} = \mathsf{x}''$ due to an impulse source at $\mathsf{x} = \mathsf{x}'$, $t = 0$ with wind field v is equal to the concentration $c_2(t) = h^r_{\mathsf{x}''}(\mathsf{x}', t)$ at $\mathsf{x} = \mathsf{x}'$ due to an impulse source at $\mathsf{x} = \mathsf{x}''$, $t = 0$ with *reverse* wind field $-\mathsf{v}$. Of course, this fact is subject to assumptions (C.1) through (C.2) being true.

Actually, this last condition (C.2) is not necessary as long as in (C.11) we replace $\lambda(\mathsf{x}, \mathsf{v}^r)$ by $\lambda(\mathsf{x}, \mathsf{v})$ and $\mathsf{k}(\mathsf{x}, \mathsf{v}^r)$ by $\mathsf{k}(\mathsf{x}, \mathsf{v})$. Although in this case a physical interpretation is missing, the reverse reciprocal Green's function can still be computed using an exiting advective dispersion computer code with proper adjustments to how λ and k are computed as functions of wind velocity.

D. Appendix: The Mathematical Form of LODI Solutions

Suppose we consider the ensemble of particles, all of whom start at a point x' and time t', and whose paths obey the random process:

$$d\mathsf{X}(t) = \mathsf{a}(\mathsf{X}(t), t, t - t')dt + \mathsf{b}(\mathsf{X}(t), t, t - t') + d\mathsf{W}, \quad \text{(D.1)}$$

where $\mathsf{X}(t)$ is random vector in R^3 for the particle position at time t and W denotes the Wiener process. The functions a and b are defined by

$$\mathsf{a}(\mathsf{x}, t, t - t') = \mathsf{v}(\mathsf{x}, t) + \nabla \cdot \mathsf{k}(\mathsf{x}, t, t - t'), \quad \text{(D.2)}$$

$$\frac{1}{2}\mathsf{b}(\mathsf{x}, t, t - t')\mathsf{b}^t(\mathsf{x}, t, t - t') = \mathsf{k}(\mathsf{x}, t, t - t'). \quad \text{(D.3)}$$

The probability density function for the particle position distribution at time t will be denoted as $g_{\mathsf{x}'t'}(\mathsf{x}, t)$. Since t' may be considered as fixed, $g_{\mathsf{x}'t'}(\mathsf{x}, t)$ can be shown to satisfy the following PDE

$$\frac{\partial g_{\mathsf{x}'t'}(\mathsf{x}, t)}{\partial t} + \nabla \cdot [\mathsf{v}(\mathsf{x}, t) g_{\mathsf{x}'t'}(\mathsf{x}, t)] - \nabla \cdot \mathsf{k}(\mathsf{x}, t, t - t')\nabla g_{\mathsf{x}'t'}(\mathsf{x}, t) = \delta(\mathsf{x} - \mathsf{x}')\delta(t - t'). \quad \text{(D.4)}$$

To solve a problem with a general source term function $s(\mathsf{x}', t')$, LODI generates an ensemble of particles starting from various values of x' and t' over which $s(\mathsf{x}', t')$ is defined, with the number of particles generated proportional to $s(\mathsf{x}', t')$. We may discretize the source over space-time voxels, so that the particles populations are subdivided into subpopulations that begin in some voxel $\mathcal{U}_{\mathsf{x}',t'}$ centered at x', t'. As the source discretization becomes finer and finer and the number of particles is correspondingly increased (so that there is a sufficient number of particles generated at each voxel), each subpopulation generated at $\mathcal{U}_{\mathsf{x}',t'}$ will be samples from a probability distribution whose density approaches the $g_{\mathsf{x}'t'}(\mathsf{x},t)$ defined above.

Because the particles are statistically independent, the density of all of the particles will be the superposition of the $g_{\mathsf{x}'t'}(\mathsf{x},t)$ with coefficient equal to the source strength at x', t'. Hence, the concentration as computed by LODI, in the limit of an infinite number of particles, for a piecewise-constant point source is given by

$$c(\mathsf{x},t) = \sum_{j,t'_j<t} g_{\mathsf{x}'t'_j}(\mathsf{x},t)s(t'_j)\Delta t'_j, \tag{D.5}$$

where t'_j is the time of the j-th packet release (with $t'_j < t$) and x' is the location of the point source. For a continuous source $s(\mathsf{x}',t')$ the concentration solved by LODI is given by

$$c(\mathsf{x},t) = \int_0^t \int_\Omega g_{\mathsf{x}'t'}(\mathsf{x},t)s(\mathsf{x}',t')\, d\mathsf{x}'dt'. \tag{D.6}$$

E. Appendix: Reciprocity Law for Basis Functions of Advection-Dispersion Equations with Memory

Define the inner product:

$$< \phi(\mathsf{x},t), \psi(\mathsf{x},t) >\equiv \int_\Omega \int_{-\infty}^{\infty} \phi(\mathsf{x},t)\psi(\mathsf{x},t)\, dtd\mathsf{x}. \tag{E.1}$$

We multiply both sides of (3.7) by $g_{\mathsf{x}'t'}(\mathsf{x},t)$ and integrate with respect to x and t, to obtain

$$\begin{aligned} &-\Big\langle g_{\mathsf{x}'t'}(\mathsf{x},t),\ \frac{\partial h_{\mathsf{x}''t''t'}(\mathsf{x},t)}{\partial t}\Big\rangle - \Big\langle g_{\mathsf{x}'t'}(\mathsf{x},t),\ \nabla\cdot[h_{\mathsf{x}''t''t'}(\mathsf{x},t)\mathsf{v}(\mathsf{x},t)]\Big\rangle \\ &\quad -\Big\langle g_{\mathsf{x}'t'}(\mathsf{x},t),\ \nabla\cdot \mathsf{k}(\mathsf{x},t,t-t')\nabla h_{\mathsf{x}''t''t'}(\mathsf{x},t)\Big\rangle = g_{\mathsf{x}'t'}(\mathsf{x}'',t''). \end{aligned} \tag{E.2}$$

Using the incompressibility assumption:

$$\nabla\cdot \mathsf{v}(\mathsf{x},t) = 0, \tag{E.3}$$

and the various identities one has

$$\begin{aligned} &\Big\langle \frac{\partial g_{\mathsf{x}'t'}(\mathsf{x},t)}{\partial t},\ h_{\mathsf{x}''t''t'}(\mathsf{x},t)\Big\rangle + \Big\langle \nabla\cdot[g_{\mathsf{x}'t'}(\mathsf{x},t)\mathsf{v}(\mathsf{x},t)],\ h_{\mathsf{x}''t''t'}(\mathsf{x},t)\Big\rangle \\ &\quad -\Big\langle \nabla\cdot \mathsf{k}(\mathsf{x},t,t-t')\nabla g_{\mathsf{x}'t'}(\mathsf{x},t),\ h_{\mathsf{x}''t''t'}(\mathsf{x},t)\Big\rangle = g_{\mathsf{x}'t'}(\mathsf{x}'',t''). \end{aligned} \tag{E.4}$$

Now, by using (3.4), we have that

$$h_{\mathsf{x}''t''t'}(\mathsf{x}', t') = g_{\mathsf{x}'t'}(\mathsf{x}'', t''), \quad t' < t''. \tag{E.5}$$